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Chapter 1

Volume Holography: Novel Materials, Methods and Applications

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Additional information is available at the end of the chapter

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Abstract

This chapter aims to establish a link between material compositions, analytical methods and advanced applications for volume holography. It provides basics on volume holography, serving as a compendium on volume holographic grating formation, specific material requirements for volume holography and diffractive properties of the different types of volume holographic gratings. The particular significance of three-dimensional optical structuring for the final optical functionality is highlighted. In this context, the interrelation between function and structure of volume holograms is investigated with view to research on and development of novel materials, methods and applications. Particular emphasis will be placed on analytical methods, assuming that they provide access for a deeper understanding of volume holographic grating formation, which appears to be prerequisite for the design of novel material systems for advanced applications.

Keywords: volume holography, Bragg gratings, photonic crystals, refractive index contrast, photosensitive materials, polymers

1. Introduction

Impressive diffraction phenomena can be found in animate and inanimate nature: in the form of iridescence in birds, insects, shells, plants or muscle cells as well as in clouds and minerals. The fascination emanating from phenomena where light interacts with micro- and nanostructures may be traced back to the principle *from structure to function*, which is not limited to optical phenomena, but can be found in nature as a fundamental quality. In case of holography, light itself is capable to create the structures with which it subsequently interacts. The (optical) functionality of a holographic structure consists in its diffractive properties.

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In turn, the principle *from structure to function* also comprises the possibility to understand function through structure. Taking advantage of this relation between structure and function culminates in the attempt of mimicking nature. Notable examples are functional surfaces with hierarchical structures based on the lotus effect to induce superhydrophobicity, the gecko effect for controlled adhesion or the moth's eye effect for anti-reflection coatings.

While the examples mentioned above remain limited to surface phenomena, the third dimension opens up entirely new possibilities with major relevance for many applications. Structures with a periodic modulation of the refractive index are of interest wherever light must be manipulated. Volume holographic gratings can be considered as such three-dimensional (3D) optical structures with diffractive properties.

Volume gratings made by nature can be found in the form of crystals wherever atoms are regularly arranged [1]. The dimensions of atomic and molecular structures usually result in interaction rather with a non-visible range of the electromagnetic spectrum, enabling access by means of X-ray crystallography. However, light-based photonic crystals (PCs), with functionality in the visible range, can be created artificially [2].

While holography allows three-dimensional imaging, the holographic structure itself extends not necessarily in three dimensions. Depending on the hologram formation technique as well as on the recording medium, the hologram itself takes shape as a surface pattern or rather emerges as a three-dimensional structure. A volume hologram or photonic crystal may only be formed if recording technique and recording medium allow modification of the optical properties in all three dimensions. The performance of such a grating with thickness in the range of 100 μm differ significantly from thin gratings or surface gratings: Volume Bragg gratings stand out due to their high diffraction efficiency, rigorous wavelength selectivity and the ability that multiple holograms may be superimposed by means of multiplexing.

There are many ways to create optical surface patterns by photolithography, self-assembly or other nano- and microfabrication methods with both, bottom-up and top-down approaches. However, entering the third dimension in optical structuring is accompanied with considerable challenges. Among existing techniques for three-dimensional optical structuring, such as direct laser writing [3] or self-assembly [4], volume holography provides the unique possibilities to create optical structures through the entire volume beyond a point-by-point, line-by-line or plane-by-plane fabrication, with high resolution and accuracy in a single step.

At the same time, the analysis of volume holographic structures emerges as a challenging task. Optical structures inside a volume may not readily be mapped by means of common microscopic methods [5]. This is where the mutuality of function and structure opens up new possibilities. In fact, the diffraction efficiency represents the only accessible parameter to entirely characterize a volume grating. Based on the optical functionality, conclusions may be drawn on grating parameters as well as on material parameters such as material response and energetic sensitivity.

Within this chapter, the interrelation between material compositions, analytical methods and advanced applications for volume holographic systems is investigated with particular emphasis on analytical methods. According to the leading idea of a correlation between function and structure, the deeper understanding of volume holographic grating formation appears prerequisite to design novel material systems for advanced applications.

2. Volume hologram formation in photosensitive materials

The mechanism of volume hologram formation in photosensitive materials, a complex process where several components are involved, is starting from the interference exposure. Holographic recording induces a generally three-dimensional spatial modulation in the optical properties. The final grating features optical functionality that consists in specific diffraction of light, to be characterized by means of its diffraction efficiency (defined as the ratio of the input readout power to the diffracted power), as well as angular response and frequency response. Different kinds of gratings are formed, according to the specific recording conditions. While the geometry clearly determines dimensionality and size of the grating, many factors influence how the material responds to light during the holographic exposure. The material response strongly depends on intrinsic material parameters, such as material composition or viscosity as well as on recording parameters, such as exposure duration and recording intensity [6].

2.1. Grating formation

Volume holographic grating formation can be attributed to different physico-chemical material transformations, depending on the type of photosensitive material. In case of polymers, an interplay of polymerization and diffusion, induced by the spatially modulated exposure, is responsible for hologram formation [7]. A light pattern is projected into the photosensitive medium, inducing local polymerization, proportional to the light intensity. Thereupon, a chemical gradient is induced, resulting in monomer diffusion and subsequent polymerization. As a consequence, the hologram is formed as a periodic modulation of optical properties, according to the recording light pattern. This grating formation mechanism is illustrated in **Figure 1**.

A special characteristic of volume gratings is that their optical functionality can be attributed to very small modulations of optical properties inside the holographic material. In case of phase gratings with a layer thickness of 200 μm, a refractive index contrast in the order of only 10^{-3} already results in diffraction efficiency close to 100%.

The contrast of such optical structures can be further enhanced with additives such as nanoparticles or quantum dots [8, 9]. It can also be combined with other mechanisms such as photochemical isomerization of optically anisotropic components or with polymer-dispersed liquid crystals (PDLCs) for switchable or tuneable optical devices [10, 11]. Furthermore, the light-induced mass transport may result in the formation of additional surface-relief gratings [12].

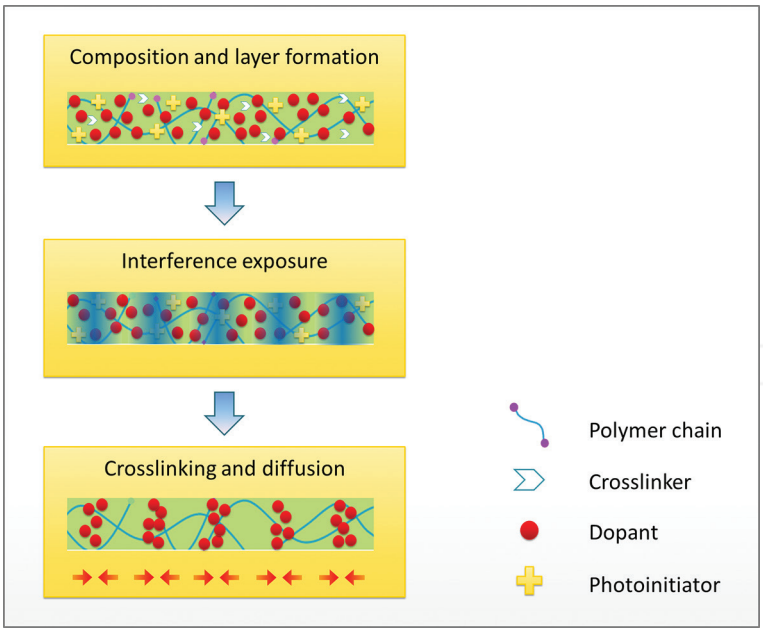

Figure 1. Schematic illustration of grating formation in photosensitive polymers: Unreacted polymer chains, crosslinker, dopant (e.g., monomers) and photoinitiator are needed for the material composition and layer formation (top). Interference exposure induces local polymerization (middle) and subsequent diffusion of components (either into the bright regions, as illustrated above, or into the dark regions) to form the permanent grating (below).

2.1.1. Specific material requirements

First of all, a photosensitive medium for volume holographic recording must be capable to undergo molecular or structural transformations with the result of a local, and in most cases, permanent change of the optical properties, as described in the previous section. In addition, and with focus on the functionality of volume holographic gratings, the medium must comply with high material standards. In this context, a number of material parameters must be optimized, namely sensitivity and dynamic range, resolution, transparency and stability. The following table gives an overview of important material parameters for volume holographic recording.

Parameter	Symbol	Unit	Target value
Layer thickness	d	[μm]	>50
Refractive index contrast	Δn	[-]	>0.001
Resolution	U	[lines/mm]	>5000
Sensitivity	S	[cm²/J]	>1
Transparency	T	[%]	>0.7

Table 1. Overview on volume holographic material parameters.

Indispensable prerequisite for the material composition is the ability to form stable layers with *thickness* d in the range of at least 50 μm where a *refractive index contrast* Δn in the order of 10^{-3} can be induced.

The material *resolution* U refers to the precision within the ability of a photosensitive material to transfer the interference pattern of exposure into a permanent modulation of optical properties during the recording process, depending on the smallest structure size of the exposure pattern. It is therefore related to the precision of the final recorded structure. With the objective to fully exploit the interference pattern of the exposure beams, high material resolution is required. With regard to the maximum spatial frequency response, that is, the highest frequency inducing a permanent refractive index modulation, a value in the range of 5000–10,000 lines per mm is aspired [13].

The *sensitivity* S provides information on how the input energy is converted into a certain holographic contrast. High sensitivity is required with respect to the possible application of low-power laser sources but also to ensure high recording speed [14]. In this context, the *dynamic range* M# refers to the total response of a medium, when divided up to n holograms. It determines how many holograms can be multiplexed in a single volume [15].

Highest *transparency* T of the material at the operating wavelength is required to achieve high diffraction efficiency. Low attenuation becomes particularly important in case of thick layers, desired for volume holographic applications. In general, losses arising from absorption and scattering should not exceed 30%. Although much lower values might be required, depending on the layer thickness, for certain specific applications [16].

Next to a high sensitivity, high spatial resolution and low losses, a high thermal stability and long-time stability of samples and systems are required [14].

Altogether, the diversity of material requirements is accompanied by a diversity of approaches to meet these needs.

2.1.2. Photosensitive media for volume holography

The range of photosensitive media used for volume holographic recording is as diverse as the spectrum of potential applications.

Although photographic emulsion, the original holographic recording material, is capable to form amplitude as well as phase holograms with good sensitivity and stability as well as high spatial resolution, it nevertheless appears inappropriate for volume holography [1]. This is due to the fact that the sample thickness is limited to only a few microns, a serious disadvantage in view of the aspired recording of thick gratings for volume holography.

Photosensitive polymers have been used as holographic media since 1969 [17]. Polymers combine many advantages, namely low cost, ease of fabrication, flexibility and the ability to be integrated in more complex systems, such as optical circuits. They fulfill the requirements for volume holographic recording with no need for solvent processing, good dimensional stability, variable thickness, high energetic sensitivity, large dynamic range and sharp angular selectivity [6, 15, 18, 19].

Among polymers for volume holographic recording, there are two material classes, differing in the mechanism of polymerization. The performances of free-radical (FRP) and cationic ring-opening polymerization (CROP) systems differ in many respects. Ranking among well-known FRP systems, glass-like polymer based on poly-(methyl methacrylate) (PMMA) with distributed phenanthrenequinone (PQ) is known as effective and thermally stable holographic recording material. Results on volume gratings within this chapter are based on investigations on gratings in free-surface epoxy-based polymer samples, prepared by microresist technology GmbH. High performance is achieved in volumetrically stable, free-surface samples with variable layer thickness [6]. The corresponding mechanism of polymerization is a cationic ring-opening polymerization. Similar to polyvinylalcohol/acrylamide (PVA/AA) material, grating formation occurs primarily as a consequence of photopolymerization and mass transport processes [20].

Photorefractive materials, such as lithium niobate, barium titanate or gallium arsenide, are capable to form temporary, erasable holograms as a result of a nonlinear optical effect [21]. Recorded data may be erased by flooding the crystal with uniform illumination. Improved properties with respect to the dynamic range, sensitivity and signal-to-noise-ratio have been demonstrated [22]. Drawbacks are partially slow response time and low stability.

2.2. Grating types

Volume holographic gratings can be categorized according to different criteria. The following section gives an overview on the different types of gratings, and how they can be distinguished with regard to their optical functionality such as diffraction efficiency and angular response.

2.2.1. Modulation

With regard to the modulated optical property, phase gratings can be differentiated from absorption gratings. Depending on the physico-chemical processes involved in the grating formation, the diffraction efficiency can be attributed to a refractive index contrast and/or to a modulation of the absorption, respectively. However, both can also be observed together [23]. In this case, the modulation of the refractive index yields a part of the total diffraction efficiency while the absorption modulation induces additional diffraction. As a consequence, it is not possible to distinguish between phase and absorption gratings, based on the diffraction efficiency. Nor can such information be derived from a microscopic image. However, it may be provided from local analysis of the optical properties, namely refractive index and absorption, respectively.

2.2.2. Dimensionality

The dimensionality of a volume holographic grating indicates in how many spatial directions the modulation spreads. This is determined by the recording interference pattern. **Figure 2** schematically illustrates how the dimensionality of a volume grating relates to the number and orientation of corresponding recording beams.

In case of a two-beam exposure, a one-dimensional volume grating is formed, illustrated by the grating planes on the left side of **Figure 2**. The grating vector $\vec{K}$ is defined by the two

wave vectors of the recording beams (this is also illustrated in **Figure 3**). At least three recording beams are needed to build a two-dimensional grating or rather optical pillars (center of **Figure 2**). A three-dimensional grating, with a modulation in all three directions, results from at least four exposure beams (right side in **Figure 2**).

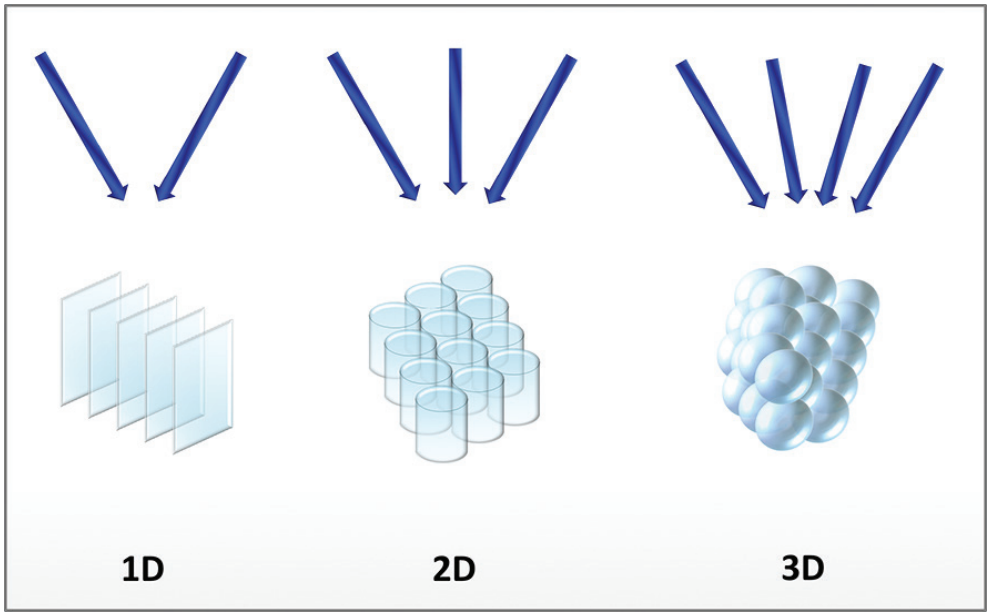

Figure 2. Schematic view of one- , two- and three-dimensional volume gratings formed by two, three and four recording beams, respectively.

Higher dimensional gratings can also be obtained by means of superposition. The same applies to the functionality of more complex structures. The diffraction pattern provides respective information according to the correlation of function and structure. A one-dimensional grating shows diffraction with only one rotational degree of freedom (left side of **Figure 2**). Each additional recording direction adds a spatial direction to the modulation of the grating with the result of a higher dimension (middle and right side of **Figure 2**).

Microscopic images of one-dimensional and three-dimensional volume phase gratings are shown in **Figure 8**.

2.2.3. Geometry

With regard to the geometry, transmission gratings can be distinguished from reflection gratings. The geometry of a grating is determined by the recording geometry, as illustrated in **Figure 3**.

The transmission and reflection curves, respectively, are strongly peaked at the Bragg angle, which is defined by Bragg's law. In case of unslanted transmission type gratings, the Bragg angle is equivalent to half the angle between reference and signal beam ($\Theta/2$). The grating period Λ is (where n is the refractive index of the recording medium and λ is the free-space recording wavelength):

$$\Lambda_t = \frac{\lambda}{2n\sin\frac{\theta}{2}} \tag{1a}$$

$$\Lambda_r = \frac{\lambda}{2n\cos\frac{\theta}{2}} \tag{1b}$$

for transmission (Λ_t) and reflection gratings (Λ_r), respectively.

A grating is transmission type if the angle between incoming light wave vector $\vec{k}$ and grating vector $\vec{K}$ is less than 90 degrees, that is, $\angle\left(\vec{k},\vec{K}\right) < \frac{\pi}{2}$. This is the case if both recording beams approach the sample from the same side (see left hand side of **Figure 3**). In contrast, the grating is reflection type if the recording beams come from both sides of the sample (see center of **Figure 3**). Again, transmission and reflection gratings may also be observed simultaneously. Beyond the possibilities to overlap gratings by means of multiplexing, superimposed holograms may also be formed due to the reflection of recording beams at the sample-substrate interface. This case of secondary gratings is illustrated on the right hand side of **Figure 3**.

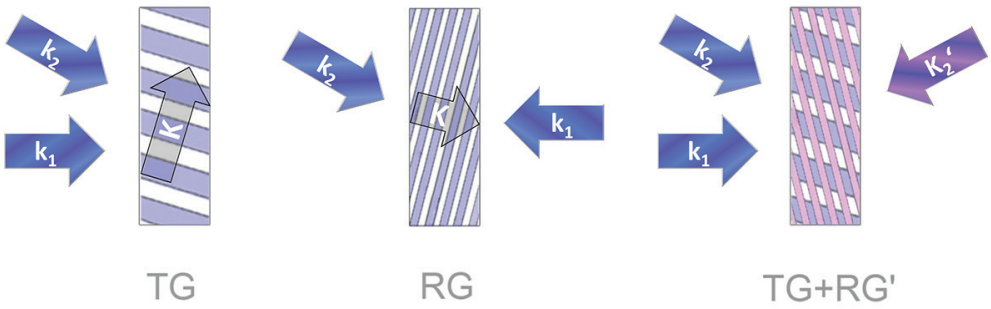

Figure 3. Recording geometries for transmission grating (TG) and reflection grating (RG). Wave vectors of recording beams $\vec{k}$ as well as grating vectors $\vec{K}$ are displayed. The reflected wave forms a secondary grating (RG') [5].

2.2.4. Selectivity

The selectivity of a volume phase grating serves as a criterion to classify the hologram with regard to the optical functionality. Therefore, it is indicated to define an important parameter with respect to the diffractive properties—the coupling constant κ:

$$\kappa = \frac{\pi\Delta n}{2\lambda} \tag{2}$$

where Δn is the refractive index contrast and λ is the recording wavelength. The coupling constant κ serves as a measure for the strength of a grating.

Holograms can be categorized into Raman-Nath type and Bragg type, respectively. A Raman-Nath hologram causes multiple diffraction orders, leading to low diffraction efficiency. A Bragg hologram shows single diffraction, enabling high diffraction efficiency and good selectivity. While Raman-Nath holograms may be recorded in a thin film, thick films are required to obtain Bragg holograms with good optical functionality [24].

With reference to the coupled wave analysis [25], the parameter:

$$\Omega = \frac{\left|\vec{K}^2\right|}{2\kappa} \tag{3}$$

with κ the coupling constant, defined by Eq. 2, can serve as an indicator for the presence of volume-type gratings. For small Ω, multi-wave diffraction occurs and little selectivity is shown. If $\Omega \geq 10$ is fulfilled, only two diffraction orders are excited [26]. This is the case if the Bragg condition is satisfied (i.e. if the probe angle corresponds to the Bragg angle, defined by Eq. 1).

From a more general perspective, a Bragg grating, also called thick grating [14], satisfies the condition:

$$d \gg \frac{\Lambda^2}{\lambda} \tag{4}$$

with d the thickness of the grating or rather the layer thickness of the recording material.

The most important two-wave case or rather the Bragg regime, is illustrated in **Figure 4**.

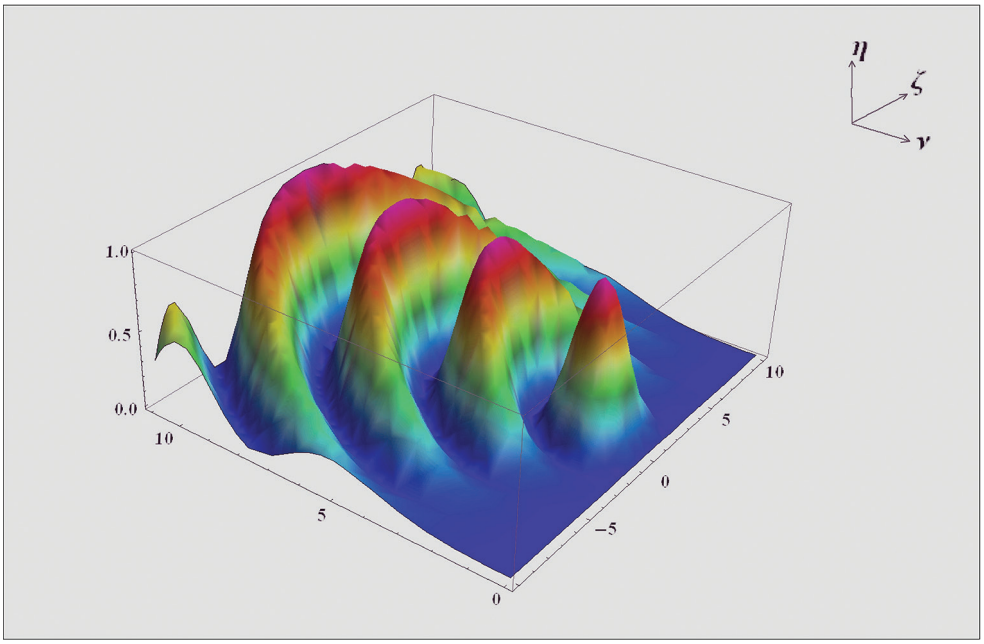

Figure 4. Diffraction efficiency η over normalized thickness ζ and normalized coupling constant ν in Bragg regime. A grating is overmodulated in case of $\nu > \frac{\pi}{2}$.

In Bragg regime, the diffraction efficiency of the first diffraction order (η) is as follows:

$$\eta_1 = sin^2(\nu) \tag{5}$$

where $\nu = \frac{\kappa d}{\cos\theta_p}$ is the normalized coupling constant with θ_p the probe beam angle (depicted in **Figure 7**).

Figure 4 shows how the diffraction efficiency distributes over normalized thickness ζ and normalized coupling constant ν. Hereby, the normalized thickness serves as off-Bragg parameter, accounting for small deviations from the Bragg condition either in terms of wavelength ($\Delta\lambda$) or in terms of angle ($\Delta\theta$) [27].

3. Analytical methods and corresponding results

The performance of volume holograms can be attributed to tiny modulations of optical properties inside the holographic material. As a consequence, direct analytical techniques, such as imaging by means of optical microscopy, are only partially appropriate for volume holograms. This is primarily due to the low optical contrast between the grating planes. But it also applies with regard to the most important three-dimensional structuring of volume holographic gratings.

3.1. Structure and function

A comprehensive analytical characterization of volume holographic gratings is possible based on the principle *from structure to function*. The idea of a correlation between structure and function for volume holograms is illustrated in **Figure 5**.

Figure 5 contrasts structural information and the related functionality for one-dimensional volume gratings. The structural information on the volume holographic grating is displayed on the left hand side. The holographic structure is described by its thickness d, the grating constant Λ and the refractive index contrast Δn. The (optical) functionality of a holographic structure consists in its diffractive properties, exemplarily displayed as angular resolved transmission on the right hand side of **Figure 5**. The link between structure and function consists in mutual determination: When the hologram is recorded, material response, exposure conditions and recording geometry determine the formation of the grating. For the final grating, the structural parameters {d, Λ, Δn} determine the diffractive properties or rather the angular resolved diffraction efficiency. In consequence, the diffraction efficiency can be utilized to access the characteristic parameters {d, Λ, Δn}. Corresponding analytical methods will be described below.

3.2. Real-time observation of grating formation

The dynamics of volume holographic grating formation may be accessed with the help of a time-resolved observation of the diffraction efficiency η(t). However, to model the grating growth and to draw conclusions on the interplay of underlying mechanisms, such as polymerization and diffusion, the time evolution of the refractive index contrast Δn(t) is needed. Potential factors, responsible for the change of the refractive index in the course of polymerization, are molecular polarizability, density and molar mass [28].

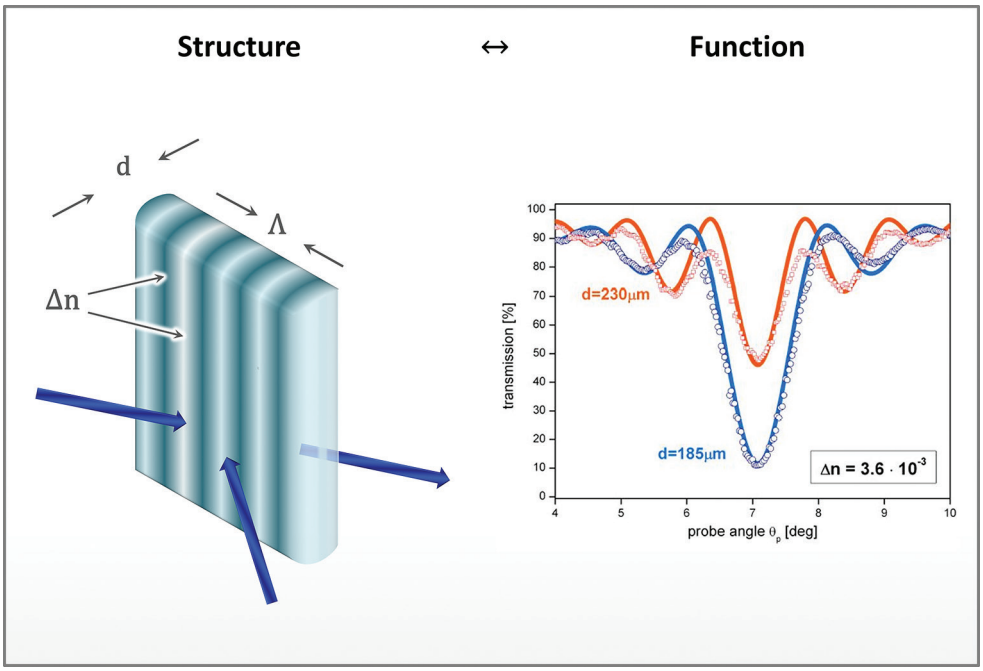

Figure 5. Relationship of structure and function for a volume holographic grating: The grating is determined by its thickness d, grating constant Λ and refractive index contrast Δn (left hand side). Those parameters determine the diffractive properties, exemplarily displayed as angular-resolved transmission (right hand side, from Ref. [6]).

Optical methods provide an elegant approach to study the kinetics of polymerization and diffusion in the course of grating formation [19]. Corresponding analysis setups feature in-situ parts for real-time, non-disturbing observation of the grating formation process and enable monitoring of the time evolution of the diffracted part of a probe beam from the very start of exposure. As a consequence, grating growth curves can be obtained.

Figure 6 shows recording and analysis setup plus corresponding grating growth curve. In this case, the growth curve reveals a transition of the refractive index contrast [29].

Figure 6 shows holographic exposure, performed by two freely propagating, s-polarized recording beams, 2 mm in diameter. Symmetric recording geometry results in unslanted gratings with periodicity of $\Lambda \approx 2\ \mu m$.

To ensure non-disturbing observation of the grating formation process, the wavelength for in-situ observation must be chosen outside of the absorption spectrum of the photosensitizer dye. In the above example, a fiber-guided 633 nm HeNe laser was used in combination with an adjustable collimator, allowing to probe with a slightly focused beam. This enables to steadily ensure a stable on-Bragg condition according to Eq. 1. A position sensitive device (PSD) was implemented to detect the diffracted light. The PSD provides time-resolved information on

the diffraction efficiency as well as on the Bragg angle. The time-resolved information on the grating constant is derived from the position of the diffracted beam on the detector. This also enables to draw conclusions on time-resolved optical shrinkage [30, 31].

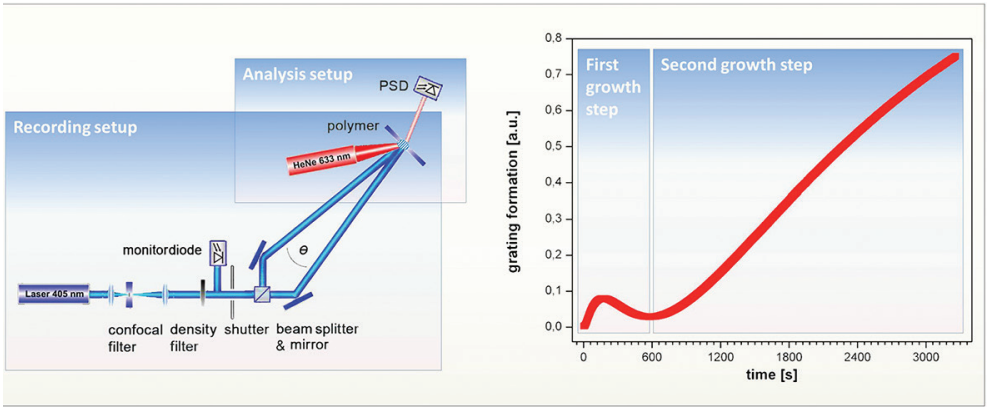

Figure 6. Recording and analysis setup for real-time observation of volume holographic grating formation: Holographic exposure is performed by two freely propagating, s-polarized recording beams with $\lambda = 405$ nm (left). The corresponding grating growth curve shows a two-step grating growth (right).

The grating growth curve shown in **Figure 6** belongs to a one-dimensional, plane-wave volume hologram of transmission type, recorded in epoxy-based polymer [29]. The characteristic two-step growth can be attributed to a transition of the refractive index contrast from positive to negative values, as a result of competing effects, taking place on overlapping time scales.

Investigations on the dynamics of volume holographic grating formation may also be applied to study the influence of important factors. This applies to material parameters, such as composition or viscosity, to grating parameters such as grating constant or geometry as well as to recording parameters, such as exposure duration or recording intensity [30].

3.3. Angular-resolved analysis

In the context of the leading idea *from structure to function*, the angular resolved analysis of volume holograms is of particular importance. Analysis of diffracted light provides basic access to the characteristic properties of the patterns causing the diffraction. Particularly, the angular-resolved diffraction efficiency provides information on the key features, such as grating constant, grating slant and, by comparison with coupled-wave-theory (RCWA) calculations, also about layer thickness, refractive index contrast and refractive index profile.

Analysis of the final holograms is usually accomplished in a rotation scan setup with collimated probe beam [32]. **Figure 7** shows a rotation scan setup plus corresponding transmission curve, that is, the angular response of a volume hologram (dots) and comparison with RCWA calculations (solid line).

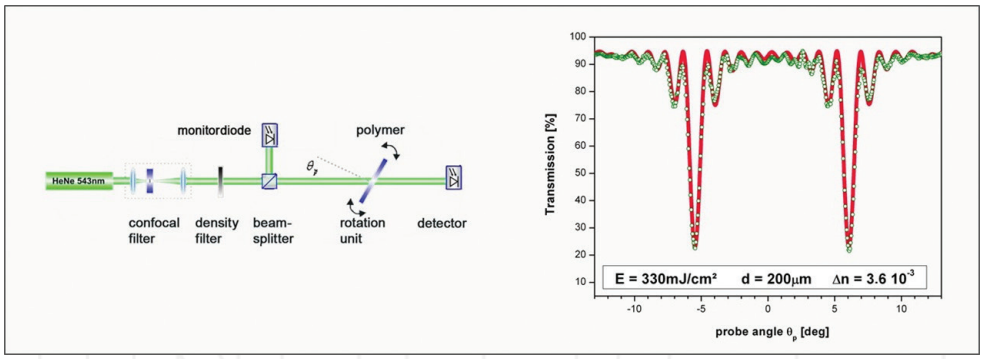

Figure 7. Rotation-scan setup (left) and corresponding angular response of a volume holographic grating, from which conclusions are drawn on grating parameters and material response (right).

The transmitted signal of a 543 nm HeNe laser is detected while the hologram under test is rotated. From the angularly resolved transmission, the following information is derived. First, the maximum diffraction efficiency can be obtained and can be correlated with the exposure energy density E to yield the material response [6]. Second, the grating constant Λ can be derived with the help of Eq. 1a. Finally, RCWA calculations can be used to derive values for the layer thickness d and the refractive index contrast Δn.

3.4. Microscopic techniques

As outlined above, low contrast is the main problem in imaging of volume holographic gratings. It might be proposed to apply fluorescent media or dyes as contrast agents to improve the image contrast. In this case, agglomeration of the contrast media along the grating planes would be prerequisite to achieve the desired effect, which cannot always be ensured. To achieve contrast in electron microscopy, conductive species are necessarily required. This is the case where nanoparticles are incorporated [33, 34]. Thus, transmission electron microscopy (TEM) is used to evaluate the degree of nanoparticle assembly [33, 35]. Scanning force microscopy (SFM) may be applied additionally, to map surface modulations [33, 36]. Nanocomposite materials are also the subject of investigations by luminescence microscopy [37]. However, mapping of nanoparticles yields only a limited description of lattice structures, not necessarily identical with the grating of interest, which is linked to the diffractive properties. In fact, it has been demonstrated that photoinsensitive nanoparticles experience counterdiffusion during grating buildup [38].

The imaging task is becoming increasingly complex without conductive species and with regard to the three dimensionality of volume phase gratings. However, it is the third dimension in particular to which the specific features of volume holograms can be assigned.

All the limitations notwithstanding, optical microscopy might nevertheless be applied to analyze volume holograms. Corresponding images are shown in **Figure 8**.

A one-dimensional volume phase grating is shown on the left side of **Figure 8**. In case of higher dimensional gratings, optical microscopy may only be applied to picture single

planes of the structure. As an example, the middle and right hand side of **Figure 8** show two planes of a three-dimensional holographic grating with hexagonal close packing crystal structure. It was produced by four mutually coherent exposure beams. The lateral distance of neighboring crystal units is 2 μm, to be read from **Figure 8**. The grating constant perpendicular to the image plane amounts to 22 μm (not shown) [39].

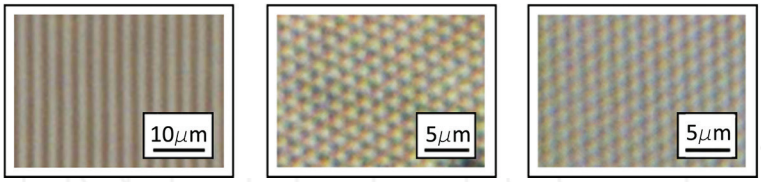

Figure 8. Optical microscopic imaging of volume phase gratings in photosensitive polymer: one-dimensional grating (left hand side) and 3D photonic grating under variation of the microscopic focal plane (middle and right hand side).

3.5. Spatially resolved diffraction analysis

Optical microscopy provides local information on the grating in the context of geometry and dimensionality. No information on the optical functionality, such as on the Bragg selectivity, is provided. However, this information is accessible according to the relation between structure and function and can be derived from spatial-resolved analysis. In case of transmission type gratings, this can be accomplished by means of scanning the lateral plane to obtain local values of the grating parameters. Local values of thickness d, refractive index contrast Δn, as well as grating period Λ and the grating slant Φ can be obtained. The lateral scan method is keeping track of the hologram shape, which is determined by the material response to the Gaussian intensity distribution of the recording beams [30].

Probing only a fraction of the exposed area is primarily for the purpose of measuring precision [29, 40]. However, it also enables scanning of the grating by moving the sample perpendicular to the optical axis. A sequence of rotation scans through the grating diameter constitutes a lateral scan. This analytical method allows the determination of the hologram characteristics along the sample surface. Thereupon, it is possible to compare and track respective properties from the center of the grating to the edges, corresponding to the areas of highest and lowest recording intensity. As a consequence, spatial sequences of the grating parameters are derived, providing insight into the local material characteristics.

Figure 9 illustrates the principle of lateral scanning. The ratio of probe beam to exposure beam diameter was 1:6. The local diffraction efficiency is displayed along the lateral position of five different volume holograms. The gratings were recorded with different exposure dose. The respective energy density of exposure (E) is displayed in **Figure 9**. The lateral scan reveals the material response, resulting in overmodulation ($\nu > \frac{\pi}{2}$) in case of $E > 330\ mJ/cm^2$.

The results from spatially resolved investigations of the grating constant reveal the influence of the recording intensity and exposure duration on the Bragg selectivity [30]. The advantage

of lateral scanning consists in the exploitation of the Gaussian intensity distribution of the recording beams. Thus, lateral scanning provides direct access to characterize the material response, based on one single exposure. As a result, other influential factors on the material response, such as pre-exposure, are eliminated.

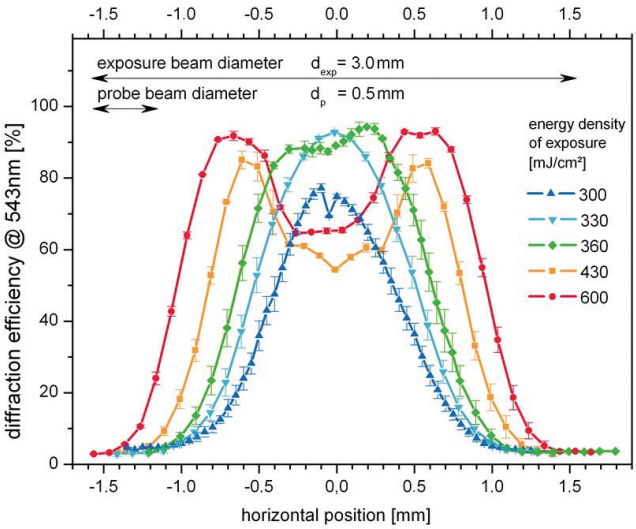

Figure 9. Principle of lateral scanning: Holograms were recorded with exposure beam diameter of 3mm and scanned step-by-step with a probe beam diameter of 0.5 mm. Five lateral scans are shown. The corresponding gratings were recorded with different exposure energy densities.

4. Volume holographic applications

The range of applications for volume holographic materials spreads wide across several disciplines. Holographic optical elements can perform the functions of mirrors, lenses, gratings or combinations of them to be applied for scanning, splitting, focusing and controlling of laser light in optical devices. The corresponding scope of applications reaches from light-guiding in general to more complex systems and operational areas, specific technologies, metrology such as holographic interferometry, through to potential use for consumer electronics such as for display technologies, as well as the many applications associated with the exploding bandwidth in meeting the demands of internet traffic and related data storage. Furthermore, holography and photolithography may arise in a powerful combination to create complex structures for micromechanical and photonic devices with potential applications not only in optics and electronics, but also in tissue engineering, cell biology and medical science as well.

Some selected application areas are described in more detail below.

4.1. Integrated optics

Photosensitive polymers are highly interesting materials for applications in integrated optics [6, 29]. This particularly applies with respect to the multi-functional applicability as well as the ability to optimize and miniaturize respective components.

Information and communication technologies are subject to a continuing shift toward optical solutions. All optical devices are needed to evade the current drawback of electro-optical chips, slowing down the whole process by forcing to work at the speed of electronics. Another objective is to reduce feature sizes in the course of miniaturization of components.

Future photonic devices such as electro-optical chips may incorporate micro-lasers and holographic optical elements (HOEs) for optical computations, interconnects and memory systems, possibly forming smaller and cheaper computer parts with higher performance [41]. Furthermore, HOEs may pave the way for the future of optical information technologies with all optical switchers, optical interconnects, (De-) multiplexers and narrow-spectral bandwidth filters, as well as photonic crystals with the potential to create integrated optical devices, capable of all optical signal processing.

The field of applications for integrated optics can further be enlarged. HOEs are also capable to efficiently redirecting light with the aim to improve light collection in solar cells [42]. Volume holographic optical elements (vHOEs) with tuneable angular and spectral Bragg selectivity, produced in instant developing photopolymer film, have recently been reported for use as lightweight, thin and flat optical elements for photovoltaic applications [43, 44].

4.2. Security technologies

Hologram encoding refers to the representation of the complex wavefield at the hologram plane, capable to encrypt information. The microscopic structure of a hologram is hard to replicate, constituting the particular applicability of holographic parts for security features. Embossed holograms are usually applied for mass production of cheap holograms for security applications, for instance on credit cards. But photopolymer holography and volume holographic parts in particular could provide considerable benefits for advanced security solutions. A volume hologram may not be copied by embossing.

Photopolymer holograms are expected to play an increasingly important role in security and authentication markets due to tight color control and strikingly realistic dimensions resulting in the unique looks of recorded images compared to embossed holograms. Photopolymer holograms can be individualized or serialized [41]. Furthermore, advanced holographic security labels could be used to fully exploit the capabilities of volume holographic systems. To further enhance the level of security, additional functionalities can be achieved by incorporation of nanoparticles. The nanoparticle-rich grating planes result in an additional security level of volume holographic labels, attained by means of specific characteristics, such as local photoluminescence in the patterned microscopic structure [37].

While two-dimensional surface holograms, currently in use, may in principle be copied by means of a point-by-point survey and imitation, the only possibility to reproduce three-

dimensional volume holograms is optical reconstruction of the original hologram and interference with the reference wave. As a consequence, the exploitation of the photosensitive volume in all three dimensions could result in a considerable increase of the security level. Recently, 2.5D nanostructures based on holographic surface-relief Bragg gratings have been demonstrated, which show tuneable diffraction in the visible spectrum and can further be combined with additional functionalities for enhanced multilevel security [45].

4.3. Biomedicine

In tissue engineering, cell biology and medical science, many applications become accessible through explicit control over molecular structure and mechanical properties, such as elasticity, cross-linking degree or surface morphology of certain biomaterials [46, 47]. In view of such applications, a combination of holographic and photolithographic processing may be used to create complex structures for micromechanical and photonic devices [2, 48]. Here again, volume holography in photosensitive polymers is of high interest for practical applications, with respect to the high flexibility and optimization ability of subsequent devices [41]. The optical functionality can be applied for the use as holographic sensors [49]. Photonic crystals are promising candidates for biosensors and bioassays. With view to the interrelation of function and structure, functional structures of different PC materials are linked with respective sensing mechanisms [50]. Optical structures have already been incorporated in hydrogels for diagnostics. Bragg grating-based hydrogel sensors as well as hydrogel microlenses are utilized as optical sensors. In both cases, a significant change in the refractive index or rather high diffraction efficiency is required to ensure good functionality [51]. Substantial benefits of holographic patterning concern the fabrication flexibility. Furthermore, hydrogels can also be structured photolithographically, taking advantage of diffusion processes. Three-dimensional structuring at the microscale results in the ability to spatially tailor biomechanical and biochemical material properties [52].

Beyond the specific material characteristics, discussed in Section 2.1.1, there are additional requirements for the composition of novel biomedical material systems, such as biocompatibility and non-toxicity [53]. Investigations on cellular behavior on one- and two-dimensional surface topography may be used to evaluate cytotoxicity and cytocompatibility as a function of mechanical properties, such as the cross-linking degree. Selective cell adhesion and spreading can be observed, depending on the (bio)chemical, physical and mechanical properties [54].

For applications in tissue engineering and medical science, the desired functionality could be achieved with a combination of optical structuring of the volume and specific modification of the surface. With view to the possibilities of miniaturization, this could result in the design of advanced biomedical implants, sensing systems and diagnostic tools for in vivo studies. In particular, intra-ocular lenses (IOLs), implanted in place of the natural eye lens in ophthalmic surgery, open up the prospect of substantial improvement, such as to overcome the systematic induction of higher order aberrations or to enhance the approximation to the original function of the natural eye lens [55, 56]. This affects not only the visual ability but also some more complex abilities of the eye such as accommodation (the capability of the eye to focus

sharply on close-up and distant objects) or brightness-darkness adaptation. With regard to the design, diffractive approaches are feasible and highly favorable in many respects compared with conventional refractive designs [57, 58]. Systems currently in use are limited to surface patterns, providing combined diffractive-refractive structures. Volume holographic systems, with potential special features involved, have not yet been applied. However, the use of refractive-diffractive optical properties by means of an integration of volume structuring, in combination with surface modification could provide highest possible functionality and applicability. Besides design, performance and functionality, biocompatibility, especially in case of foldable hydrogel lenses, must be ensured [59, 60]. Prospective IOLs could fulfill their function with an optically structured volume, leaving the surface free for other functionalities. Thus, late postoperative opacification of implanted lenses, resulting in glare and misty vision, might be addressed based on surface modifications for specific bio-interaction [60]. In any case, the integration of optical functionality into the volume of intra-ocular lenses might succeed according to the leading idea *from structure to function* and would be accompanied with considerable benefits.

5. Conclusion and outlook

In this chapter, the interrelation of structure and function for volume holographic gratings was investigated with view to materials, methods and applications for volume holography. Hereby, volume holograms were considered as three-dimensional optical structures with specific functionality in terms of diffractive properties. The mechanism of volume holographic grating formation in photosensitive polymers was described. Specific requirements for volume holographic materials and respective material systems were discussed. Different types of volume holographic gratings were characterized. Analytical methods for volume holograms were presented for the real-time observation of grating formation as well as for the analysis of the final optical functionality. In addition, imaging techniques were discussed and optical microscopy was applied to image 1D and 3D volume phase gratings. Lateral scanning was proposed to exploit the Gaussian intensity distribution of the recording beams, providing direct access to the material response, based on a single exposure. Finally, some selected application areas have been described with respect to the specific advantages of volume holographic materials for the respective applications. It could be demonstrated that the opening up of new applications for volume holography is accompanied with the design of novel, functionally tailored material systems. Therefore, a deeper understanding of volume holographic grating formation mechanisms remains required, driving the need for appropriate analytical methods. In this context, future opportunities and challenges related to the three dimensionality of volume holographic gratings have been highlighted.

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